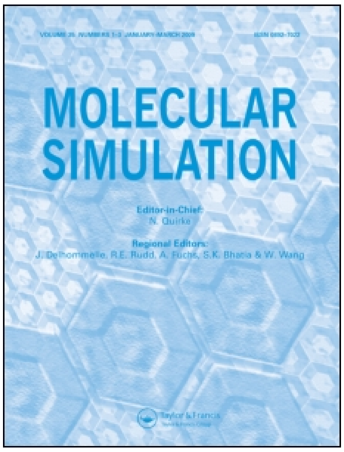

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NONLINEAR RESPONSE TO STEADY AND OSCILLATING PLANAR COUETTE FLOW

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Nonequilibrium molecular dynamics simulations were performed to determine the viscoelastic response of a two dimensional system of soft sphere disks to a combination of steady and oscillating planar Couette flow. Anomalous behaviour observed in the frequency dependent shear viscosity at high steady shear rates was found to be the result of nonlinear coupling between shear and normal stress modes. The results were compared with the predictions of the Goddard-Miller model which was found to be in qualitative agreement at low steady shear rates, but broke down at higher steady shear rates.

Keywords: NEMD; Planar Couette flow; Goddard-Miller model.

1. INTRODUCTION

Many practical problems involving oscillations can be solved satisfactorily if it is assumed that the system is linear. A system is defined as linear if the frequency of the response (ω_{resp}) coincides with the frequency of the applied field (ω_{app}). The response of a linear system to an external field can be represented by the equation, $\omega_{resp} = a\omega_{app}$. In the case of a nonlinear system, the response is nonlinear with respect to the applied field, for example [1], it might be $\omega_{resp} = a\omega_{app} + b\omega_{app}^2$. A linear oscillating system is one where the restoring force is linearly dependent on the deflection, and the damping force is linearly dependent on the velocity, resulting in a system where the output signal is in phase with the input signal. The motion of a one dimensional system undergoing linear oscillations in the x-direction can be modelled using the following equation,

$$\ddot{x} + \beta\dot{x} + \omega^2 x = E \tag{1}$$

where $\omega = \sqrt{k/m}$ and m is the mass, k the strength of the restoring force, β the damping force coefficient, and E an external field applied to the system. An example of a non-linear oscillating system is the Duffing equation,

$$\ddot{x} + \varepsilon f_D(\dot{x}) + f_R(x) = E \tag{2}$$

where, $\varepsilon \ll 1, f_R(x)$ is the non-linear restoring force and $\varepsilon f_D(\dot{x})$ is the non-linear damping force.

Unlike one-dimensional systems, multi-dimensional systems can posses the peculiar characteristic of multiple resonance frequencies. An n-dimensional system can posses as many linear natural frequencies, as there are degrees of freedom. The non-linearity of the system can cause the n modes of motion to become strongly coupled, this is termed internal resonance. For a system to show internal resonance, it would need to satisfy the condition, $m_1\omega_1 + m_2\omega_2 + \ldots + m_n\omega_n \approx 0$, where $m_1, m_2, \ldots, m_n$ are positive or negative integers [2]. For example a system with quadratic non-linearity, and internal resonance, can transfer energy between modes. If a system has the internal resonance of $\omega_2 = 2\omega_1$, a saturation phenomena exists such that, if the system is driven at $\Omega \approx \omega_2$, at small amplitudes of driving force, only the second mode is excited. If the amplitude increases and exceeds a critical value, the first mode begins to respond while the second mode is saturated. If the amplitude continues to increase the additional energy is transferred into the first mode. If this same system is driven at $\Omega = \omega_1$, under some circumstances the system will not develop a steady state and the energy is continuously exchanged between the two modes.

The classic system, displaying a coupling of the modes, is the spring pendulum. According to linear theory, the two modes of the pendulum are uncoupled, that is, motion of the pendulum in one mode is completely independent of the motion in the other mode. In 1933, Gorelick and Witt [3], determined experimentally that when $\omega_2 \approx 2\omega_1$, the two modes were indeed coupled [2]. Their experiment involved setting the pendulum at $\theta = \theta_0$, where $\theta_0 \neq 0$ and very small, while the mass m is pulled down, stretching the spring. The pendulum will oscillate up and down with spring-type motion, with the pendulum-type motion steadily increasing at the expense of the spring-type motion. After a further time span, the pendulum-type oscillations decreases with the spring-type oscillations steadily increasing. The energy is continually transferred back and forth between the two modes.

The Lagrangian for this type of system is given by the following equation,

$$L = \frac{1}{2}m\,[\dot{x}^2 + (l + x)^2\dot{\theta}^2] - \frac{1}{2}kx^2 - mg(l + x)(1 - \cos\theta) + mgx \tag{3}$$

which yields the following equations of motion,

$$\ddot{x} + \omega_2^2 x - (l - x)\dot{\theta}^2 - g\cos\theta = 0 \tag{4}$$

and

$$\ddot{\theta} + \frac{g\sin\theta + 2\dot{x}\dot{\theta}}{l + x} = 0 \tag{5}$$

where $\omega_2 = \sqrt{k/m}$. For small oscillations, the non-linear terms in the equations of motion can be ignored, giving a system with uncoupled modes. A spring-type mode of resonant frequency ω_2 and a pendulum-type mode of frequency $\omega_1 = \sqrt{g/l}$. However if the nonlinear terms are included, the two modes of the system are clearly coupled.

The macroscopic response of a fluid system to a time dependent external field can be considered in the same way as a system of coupled harmonic oscillators, or the Duffing equation. In each case we study the response of a single scalar variable (such as the shear stress) as a function of frequency or time, and we compare that with the behaviour of a simpler linear or nonlinear mechanical system. In the simulations of the fluid system performed here, we observe nonlinear behaviour, for example, by monitoring the change in the normal stress difference, $P_{xx} - P_{yy}$. From linear response theory it can be shown that the system should display no normal stress difference so this is a purely nonlinear mode. However, before looking in detail at nonlinear modes we will consider the predictions of linear response theory for this fluid.

The real and imaginary components of the frequency dependent shear viscosity can be calculated from the equations of linear response theory.

$$\eta(\omega) = \int_0^\infty dt \langle P_{xy}(t) P_{xy}(0)\rangle_0 \cos(\omega t) \tag{6}$$

$$\eta'(\omega) = \int_0^\infty dt \langle P_{xy}(t) P_{xy}(0)\rangle_0 \sin(\omega t) \tag{7}$$

This functional form of the stress-stress autocorrelation function determines the frequency dependence of the real and imaginary parts of the shear viscosity. For a two dimensional system, the stress-stress autocorrelation has a long time tail which decays as t^{-1} at long time, and this implies that,

$$\langle P_{xy}(t) P_{xy}(0)\rangle \approx A t^{-1} \tag{8}$$

Using the assumption of the long time tail for a two dimensional system and the above equations, Zwanzig [5] derived the following expression for the real component of the frequency dependent shear viscosity,

$$\eta(\omega) = -B\log\omega + O(1) \tag{9}$$

It has been shown theoretically, that $\eta(\gamma)$ for a two dimensional system experiencing a steady planar Couette flow γ, will be of the form [4],

$$\eta(\gamma) = -A\log\gamma + O(1) \tag{10}$$

The equation for the frequency dependent shear viscosity $\eta(\omega)$, has the same functional form as the steady shear dependent viscosity $\eta(\gamma)$, but with different coefficients.

The discovery of the long-time tail by Alder and Wainwright [6], and its subsequent explanation by vortex velocity fields, has given cause for the use of corotational models. This has come about because the linear response equations are not invariant under rotation, which limits the theory's ability to predict fluid behaviour at large deformations [5]. This problem with the theory can be overcome, if the vorticity of the system is removed, thus generalising the linear response equations to the system. The linear response theory can be generalised by applying the equations to a corotational model. One such model is the Goddard-Miller model.

2. GODDARD-MILLER MODEL

In applying a corotational model to the system, the system is observed from a frame of reference which moves through space relative to the fixed laboratory frame. At time $t = 0$, the origin of the corotating reference frame coincides with the origin of the laboratory frame, which also coincides with the position of a particle. The corotating frame follows the trajectory of the particle, rotating with the same angular velocity as that of the local fluid element [7]. Transforming the linear response equations to the corotating frame, the rotation of the fluid is removed with respect to the reference frame.

A two dimensional system experiencing a steady uniform shear $\gamma(t)$, will have a streaming velocity $\mathbf{u}$ satisfying the equation,

$$\mathbf{u} = \gamma y \mathbf{i} \tag{11}$$

Using linear response theory in a stationary frame, the shear stress tensor **P** is related to the shear rate tensor $\varepsilon(t) = \nabla \mathbf{u}$ by the equation,

$$\mathbf{P}(t) = \frac{V}{k_B T} \int_0^t ds \langle \mathbf{P}(s)\mathbf{P}(0) \rangle \cdot \varepsilon(t-s). \tag{12}$$

The stress-stress auto-correlation function decays as t^{-1} and so the system experiences vorticity with a corresponding angular momentum of,

$$\omega = \frac{(\nabla \times \mathbf{u})}{2} \tag{13}$$

The transformation from the stationary reference frame to the co-rotating frame is described by the transformation matrix Ω which satisfies the condition,

$$\frac{d}{dt}\Lambda = \omega \times \Omega = \Omega \cdot \Lambda \tag{14}$$

The transformation matrix relates any tensor $\Lambda(t)$ to an equivalent tensor Λ^c (t) in the corotating frame by the expression,

$$\Lambda(t) = \exp(t\Omega) \cdot \Lambda^c \cdot \exp(-t\Omega) \tag{15}$$

Applying the linear response equation to the corotating frame for the shear stress gives the equation,

$$\mathbf{P}^c(t) = \frac{V}{k_B T} \int_0^t ds \langle \mathbf{P}(s)\mathbf{P}(0) \rangle \cdot \varepsilon^c(t-s) \tag{16}$$

Transforming this equation back to the stationary frame gives the expression for the pressure tensor,

$$\mathbf{P}(t) = \frac{V}{k_B T} \int_0^t ds \langle \mathbf{P}(s)\mathbf{P}(0) \rangle \cdot \exp(s\Omega)\varepsilon^c(t-s)\exp(-s\Omega) \tag{17}$$

Taking the xy components of the pressure and strain tensors and working out the relevant components of the transformation matrix, the resulting expression for the shear viscosity becomes,

$$\eta(\gamma) = \frac{V}{k_B T}\int_0^{\infty} dt \langle P_{xy}(t) P_{xy}(0) \rangle \cos(\gamma t) \tag{18}$$

This expression for the shear dependent viscosity has the same functional form as the frequency dependent shear viscosity, $\eta'(\omega)$ given in equation (8). Unfortunately the Goddard Miller model is a somewhat rough approximation. The gradient of the streaming velocity, $\nabla\mathbf{u}$ is composed of two components, a symmetric component $\varepsilon/2$ and an antisymmetric component $\Omega/2$ [5]. The Goddard Miller model only considers the antisymmetric component, in the transformations.

3. SIMULATION DETAILS

The time dependent SLLOD equations of motion [8] are applied to a two dimensional system of 896 soft-spheres. The pair interaction potential between two soft-spheres is of the form $\phi = \varepsilon(\sigma/r)^{12}$, inside a cut-off radius of 1.5 σ. Out side this cut-off the interaction potential is zero. Reduced units are used throughout, where distances are in units of σ, where σ is the diameter of the particle, and the temperature is measured in units of ε/k_B, where ε is the depth of the Lennard-Jones energy potential, and k_B is Boltzmann's constant. In all simulations the density was set at $\rho = 0.9238$, and the temperature maintained at a value of 1.0.

The shear rate applied to the system was composed of a combination of a strong steady shear and a smaller oscillating shear. Therefore,

$$\gamma(t) = \gamma_0 + \gamma_1 \cos(\omega t) \tag{19}$$

For fixed values of the steady shear rate γ_0, the amplitude of the oscillating component was held constant at $\gamma_1 = 0.1$ and its frequency varied. The response function of primary interest was the frequency dependent shear viscosity which was monitored over the same range of frequencies, typically $0 < \omega < 30$. The fixed values of the steady strain rate chosen were $\gamma_0 = 0.5$, 1.0, 1.5 & 2.0. In this way we were able to estimate the coupling of the steady component of the strain rate to the frequency dependent response. We have not attempted to examine the reverse effect in these simulations.

The SLLOD equations of motion are given by

$$\dot{\mathbf{q}}_i = \frac{\mathbf{p}_i}{m_i} + \mathbf{i}\gamma(t)y_i \tag{20}$$

$$\dot{\mathbf{p}}_i = \mathbf{F}_i - \mathbf{i}\gamma(t)p_{yi} - \alpha\mathbf{p}_i \tag{21}$$

where α is the thermostatting multiplier whose instantaneous value is given by

$$\alpha = \frac{\sum_i \mathbf{F}_i \cdot \mathbf{p}_i - \gamma(t)\sum_i p_{xi}p_{yi}}{\sum_i(\mathbf{p}_i^2/m_i)} \tag{22}$$

With the momentum of each particle defined as a peculiar momentum, the temperature of the system is defined by the equation,

$$\frac{2Nk_BT}{2} = \frac{1}{2m}\sum \mathbf{p}_i^2 \tag{23}$$

When a particle's y coordinate moves outside the primitive cell, the value of its velocity in the x direction needs to be calculated using equation (20). This is a little more complicated than the usual time independent case.

With the system driven by a time dependent force of frequency ω it is assumed that the response of the shear stress, $P_{xy}(t)$ will also be periodic with the same period $T = 2\pi/\omega$. This makes it possible to expand the measured response $P_{xy}(t)$ in a Fourier series so that [9],

$$P_{xy}(t) = \frac{1}{2}a_0 + \sum_{n=1}^{\infty}[a_n\cos(n\omega t) + b_n\sin(n\omega t)] \tag{24}$$

where the Fourier coefficients are given by

$$a_n = \frac{2}{T}\int_0^T dt P_{xy}(t)\cos(n\omega t) \tag{25}$$

$$b_n = \frac{2}{T}\int_0^T dt P_{xy}(t)\sin(n\omega t) \tag{26}$$

From linear response theory, the phase variable $P_{xy}(t)$, of a system undergoing a time dependent shear is given by the equation,

$$P_{xy}(t) = -\int_0^t ds\chi(t-s)\chi(s) \tag{27}$$

where $\chi(t) = \beta V\langle P_{xy}(t)P_{xy}(0)\rangle$. The right hand side of equation (27) is a convolution, hence in the frequency domain,

$$P_{xy}(\omega) = -\chi(\omega)\gamma(\omega) \tag{28}$$

this suggests that a system driven at a frequency ω will respond only at a frequency ω. This is not all together true in practice, the non-linear response of the system will contain contributions from the higher harmonics of the driving frequency. With $P_{xy}(t)$ as the response function, the real part of the complex susceptibility $\chi(\omega)$ is the shear viscosity. In NEMD dynamics the system is driven at a frequency ω and the response of $P_{xy}(t)$ is monitored at the same frequency with the frequency dependent shear viscosity being calculated from equation (28).

Another method of calculating the frequency dependent shear viscocity is to consider the energy dissipation of the system. The total internal energy of the system is given by the equation,

$$H_0 = \frac{1}{2m}\sum_i p_i^2 + \phi \tag{29}$$

Differentiating this equation with respect to time, and substituting the equations of motion (equations 20 and 21), the energy dissipation is given by

$$\dot{H}_0(t) = -P_{xy}(t)\,V\gamma(t) - 2Nk_BT\alpha(t) \tag{30}$$

If the system is in a steady state the integral of $\dot{H}_0(t)$ over one period of the external field must be zero. Therefore, the real part of the frequency dependent shear viscosity is given by

$$\mathrm{Re}[\eta(\omega)] = \frac{4\rho k_B T\langle\alpha\rangle}{\gamma'^2} \tag{31}$$

This gives a second route to calculate the frequency dependent shear viscosity.

4. OVERVIEW OF PREVIOUS RESULTS

In 1983, Evans and Morriss [13], reported the results of computer simulations of two-dimensional systems of 896 and 3584 particle undergoing steady shear. The simulations were performed at the three different densities of $\rho\sigma^2 = 0.96$, 0.9238, and 0.6928. Their results did not agree with Kawasaki and Gunton [4] who predicted that the shear viscosity $\eta(\gamma)$, would be linear in $\log(\gamma)$, for all values of γ. The simulations performed by Evans and Morriss showed a "turnover regime" at small strain rate, where the shear viscosity is essentially independent of the strain rate. At higher strain rates, the shear viscosity is linear in $\log(\gamma)$, in agreement with the theoretical predictions. They found that reducing the density of the system increased the point of transition from the "turnover regime" to the region of logarithmic variation. They concluded that the discrepancy between the simulations and the theoretical predictions were caused by the development of fluctuating convective cells within the system. When measures were taken to remove the kinks in the velocity profile and maintain the linear velocity profile assumed in the theory, the results of the simulations agreed with the theoretical predictions. The results also showed that changing the density and system size did not change the qualitative characteristics of the system.

In 1985, the same group [9], presented the results of computer simulations of viscoelasticity in two-dimensional fluids. An oscillating strain rate was applied to a two-dimensional 896 particle, soft disk system of the form,

$$\gamma(t) = \gamma_1 \cos(\omega t) \tag{32}$$

with the system density maintained at $\rho\sigma^2 = 0.9238$. The simulations were performed for a range of frequencies, at two different amplitudes, $\gamma_1 = 0.1$ and $\gamma_1 = 1.0$. The results showed that the viscosity was logarithmic in the frequency of the strain rate, except at low frequencies, where a "turnover regime" was observed in which the shear viscocity was independent of the frequency. This was consistent with their previous results for steady shear flow and suggested that the "turnover regime" exists for both small strain rate and small frequency. This consistency in the results prompted them to look more closely at the Goddard-Miller model that suggests that the frequency and strain-rate dependence of the shear viscosity should have the same functional form and the same coefficients. Although the functional forms are the same over an intermediate range of both strain rate and frequency, the coefficients are quite different.

The results also showed super-harmonic response. At a strain rate of 1.0, the shear stress, responded strongly at a frequency of 3ω, and the pressure responded strongly at a frequency of 2ω. This super-harmonic response contradicts the assumption of linear response for the system (that is, that a system driven at a frequency ω will only respond at the same frequency). Super-harmonic response is a purely nonlinear effect.

5. RESULTS AND DISCUSSION

5.1. Frequency Dependent Response

The simulations performed here differed from those of previous computer experiments [6,13], in that we explore the nonlinear coupling of a steady strain rate to the frequency dependent response to an oscillating strain rate. Figure 1 presents the real and imaginary components of the frequency dependent response of the shear viscosity as a function of the logarithm of the frequency, for each of the four values of the steady strain rate, $\gamma_0 = 0.5$, 1.0, 1.5 & 2.0, respectively. At zero frequency, the system behaves as a shear thinning non-Newtonian fluid where the viscosity decreases consistently with increasing steady shear rate. At low frequencies, only the graphs for $\gamma_0 = 0.5$ and $\gamma_0 = 1.0$ showed any discernible "turnover" as seen in previous results [6,13] where $\eta(\omega)$ is independent of the frequency. This result would seem to support the suggestion of Morriss and Evans [6] that $\eta(\gamma,\omega)$ is independent of γ and ω when both are small. The system experienced shear thickening (in frequency rather than strain rate) at low frequencies for all the strain rates tested, with the degree of shear thickening increasing with increasing steady strain rate, peaking, and then shear thinning rapidly with increasing frequency. The difference in the shear viscosity for a given frequency at a given strain rate diminished with increasing frequency to the point where there was no discernible difference in the shear viscosities at the highest frequencies.

The most striking feature of the graphs is the progressive change of the peak in the real component of the shear viscosity with increasing steady shear rate. The peak becomes progressively flatter with increasing steady shear culminating in a sharp trough at a frequency near $\omega \approx 1$, at a steady shear rate of $\gamma_0 = 2$. A similar effect can also be seen in the imaginary component. This major feature of the real part of the shear viscosity also appears in the imaginary component as well.

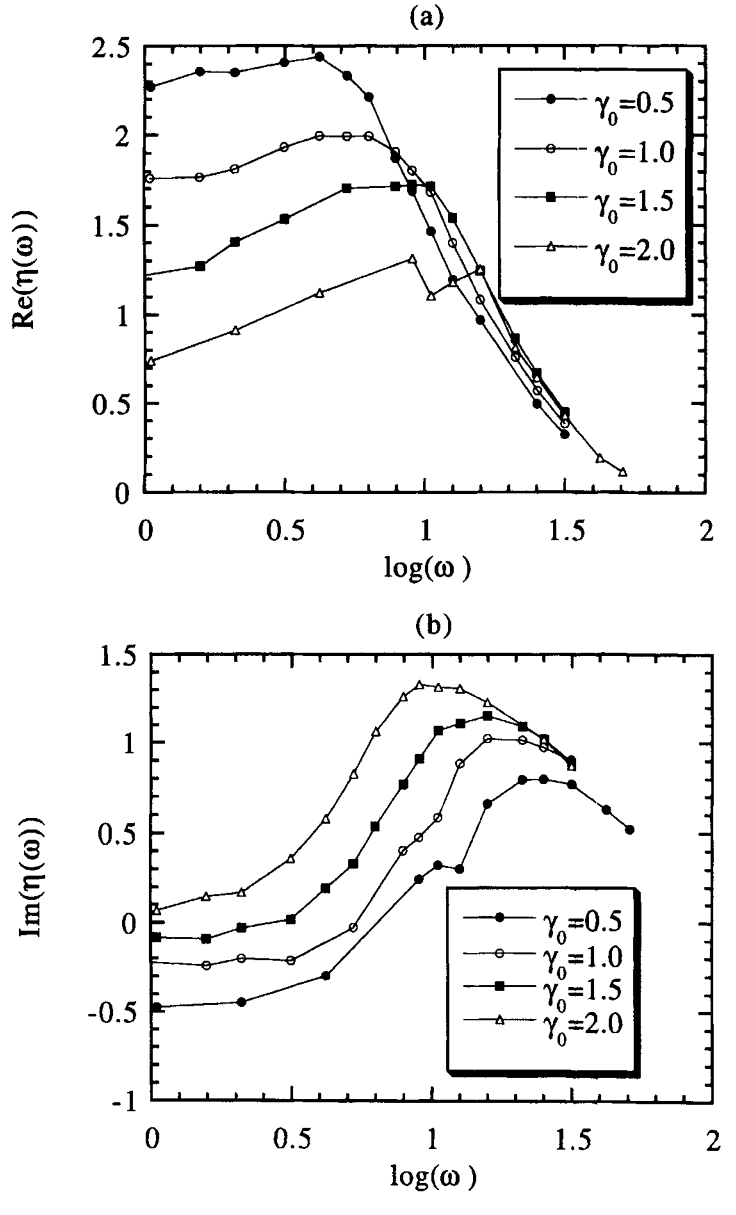

FIGURE 1 Real (a) and Imaginary (b) components of the shear viscosity for each of the shear rates, against the log of the frequency, $\gamma_0 = 0.5$, $\gamma_0 = 1.0$, $\gamma_0 = 1.5$ and $\gamma_0 = 2.0$.

5.2. Coupling of Resonance Modes

Figure 2, shows the normal stress difference, $P_{xx} - P_{yy}$ versus frequency for a system size of 896 particles, at each of the shear rates studied in this paper. The most striking features occur in the graphs corresponding to $\gamma_0 = 1.0$, $\gamma_0 = 1.5$ and $\gamma_0 = 2.0$. In each case, the normal stress difference is negative, which in rheological experiments has only been found at very high strain rate [14]. Each of these graphs show an ever deepening trough with increasing shear rate, occurring at an increasingly higher frequency range. The frequency ranges in which each of the troughs occur is closely correlated to the frequency range where the anomalous behaviour is visible in the frequency dependent shear viscosity shown in Figures 1(a) and (b). In the case of $\gamma_0 = 0.5$ there is no discernible trough. A trough may exist at a very low frequency, but its amplitude will be small and therefore not be detectable in the graphs.

The troughs in the graphs indicates that the system is experiencing a sudden rise in the pressure, perpendicular to the streaming flow. There are at least two possible explanations for the sharp rise in the perpendicular press-ure. It is possible that the system is forming an ordered structure in this narrow frequency range, or else the system was directly exciting that perpendicular mode in a narrow range of frequencies. The motivation for considering

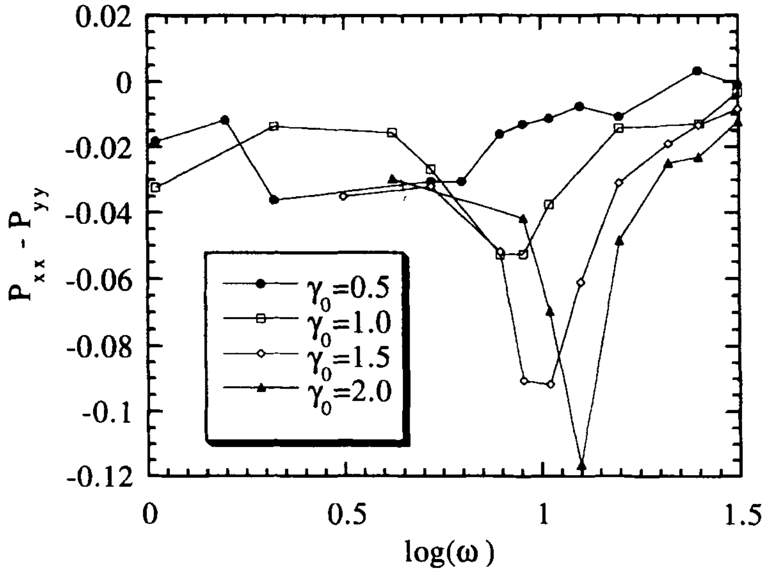

FIGURE 2 Normal stress difference for each of the four shear rates.

the possibility of an ordered structure came from the results of previous experiments where two dimensional systems had been observed to form an ordered structure (the so-called "string phase" or "necklace structure") when driven by a strong steady shear rate [10]. A divergence in the normal pressure difference is also observed with the onset of the "string phase".

To test the possibility of a change in the structure of the system, the pair distribution function $g(r)$ was calculated for steady strain rates of $\gamma_0 = 2.0$ at frequencies of $\omega = 8.976$, $\omega = 12.566$ and $\omega = 50.671$. No change in the pair distribution function was observed for the three frequencies, thus we concluded that the normal stress effect is not associated with a structural change in the fluid.

The most plausible explanation for the sharp peak in the normal stress is that a nonlinear mode is excited. This mode would resemble the saturation phenomenon observed in simple nonlinear oscillators discussed in the introduction. Considering the following graphs, Figure 3 shows the two pressure components at a steady strain rate of $\gamma_0 = 2.0$, while Figure 4 shows the two pressure components at a steady strain rate of $\gamma_0 = 0.5$. In the case of $\gamma_0 = 2.0$, the pressure in the y-direction rises sharply, suppressing some of the system response in other directions, similar to the exchange of energy

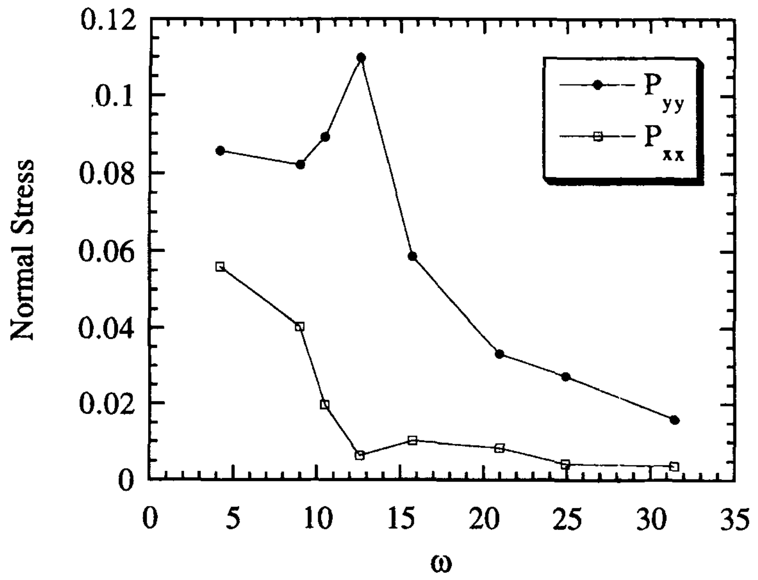

FIGURE 3 Parrallel and normal stress components at the steady strain rate of $\gamma_0 = 2.0$.

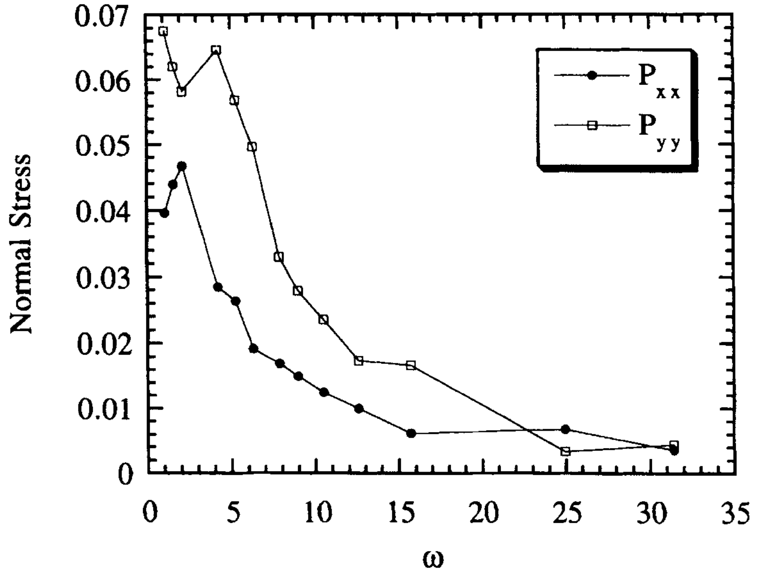

FIGURE 4 Parrallel and normal stress components at the steady strain rate of $\gamma_0 = 0.5$.

seen in the spring pendulum mentioned in the introduction. The added excitation of the system as a result of the resonance in the y-direction appears to be associated with a lowering of the shear viscosity within a narrow frequency range. The effect is not apparent when $\gamma_0 = 0.5$.

5.3. Goddard-Miller Model

The Goddard-Miller model predicts that the response of the system to the steady component of the strain rate is in some way coupled to the system response to the frequency dependent component of the strain rate. It is anticipated that for a two dimensional system experiencing a combined steady and oscillating shear, any change in the steady component will effect the shear viscosity in the same way as a corresponding change in the frequency [7,5,9]. Figure 5 shows no agreement with this prediction. Only the three lowest steady strain rates have been used for comparison, as the theory is only expected to apply to linear systems, and as the previous results show, at the highest strain rate, the system displays non-linear behaviour. In the low frequency range the system shear thickens with increasing frequency, however, increasing the steady shear results in the opposite effect, shear thinning. In the higher frequency range, the system behaviour

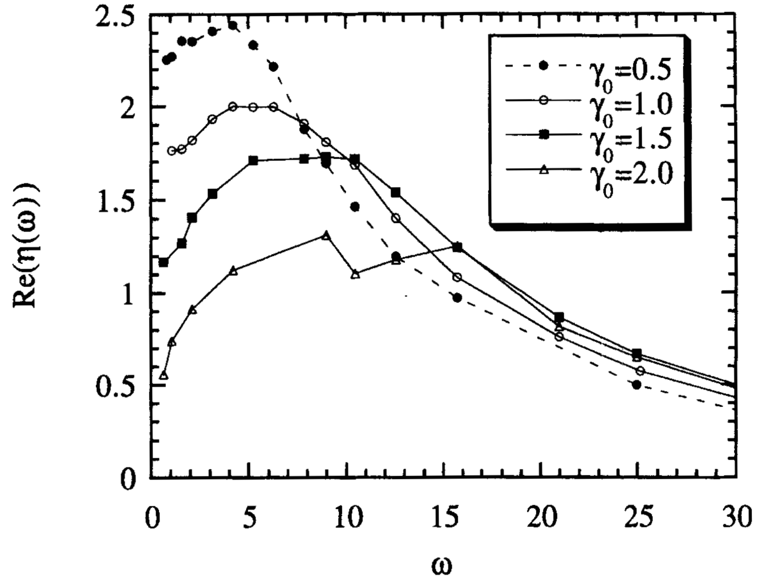

FIGURE 5 The real component of the shear viscosity for each of the four strain rates.

also disagrees with this prediction. The system experiences shear thinning with increasing frequency, wheras increasing the steady component results in shear thickening.

Figure 2, which shows the graphs of the normal stress difference $P_{xx} - P_{yy}$, should also agree with the prediction of the Goddard-Miller model. Unlike the graphs for the shear viscosity the graphs for the normal stress differences show an approximate agreement with the preidictions of the theory. The troughs in each of the four graphs occur at increasingly higher frequencies with increasing steady shear, however the shifts are not constant. The trough in the graph of $\gamma_0 = 0.5$ is not clearly visible in the low frequency noise. The troughs for $\gamma_0 = 1.0$ and $\gamma_0 = 1.5$ occur at $\omega = 8.5$ and $\omega = 9.5$ respectively. With the steady shear set at $\gamma_0 = 2.0$, the Goddard-Miller model would predict that the trough should be found at $\omega = 10.5$, however it is instead found at $\omega = 12.5$, further supporting the argument that the theory breaks down at higher strain rates.

6. CONCLUSION

The results of the simulations of an atomic fluid under a strain rate composed of a steady and a small superimposed oscillating component, showed that the frequency dependent shear viscosity was a complicated function of the frequency. There is clear evidence that the strength of the steady component of the strain rate has a large effect on the observed frequency dependent viscosity. At small frequencies the "turnover regime" has a steadily decreasing viscosity with increasing steady strain rate. At high frequency the effect of the steady component diminishes and probably disappears in the limit of large frequency. The anomalous behaviour observed in the frequency dependent shear viscosity at the highest strain rate was found to be a result of a coupling to a normal stress mode of the same frequency. The normal stress effect was visible at all but the lowest strain rate, increasing significantly with increasing steady shear. The normal stress effect appears to be a nonlinear threshold phenomena, as it became significant only at the highest steady strain rates.

The results were found to disagree with the predictions of the Goddard-Miller model, in the case of the frequency dependent shear viscosity. Due to the lack of qualitative agreement, no quantitative comparisons between the results and the Goddard-Miller predictions were attempted.

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